

In the Specification (clean copy as amended)

Kindly amend the Specification as follows:

Page 1, second full paragraph:

A1 As known, zeolite is used as a catalyst for conversion of aromatic compounds having substituent(s), for example, for xylene isomerization, toluene disproportionation, etc. It is known that zeolite, especially mordenite-type zeolite is used for conversion of aromatic compounds having at least 3 substituents (Japanese Patent Laid-Open No. 14430/1983).

Paragraph bridging pages 1 and 2:

A2 Zeolite is a porous crystal of which the pores are uniform and have a molecular-level size. It can be a catalyst having good activity and selectivity for conversion of aromatic compounds having a relatively small molecular size, for example, for xylene isomerization, toluene disproportionation or the like, and is so used in some industrial-scale plants. However, for conversion of large-size molecules, using zeolite is often problematic in that the reactant molecules could not penetrate into the zeolite pores, or even if having penetrated thereinto, they could not diffuse rapidly through the pores to receive satisfactory conversion activity. On the other hand, among many kinds of zeolite, pentacyl-type zeolite, mordenite-type zeolite, β -type zeolite, and faujasite-type zeolite are widely used.

Page 2, first full paragraph:

A3 β -type zeolite and faujasite-type zeolite have pore apertures with the largest size of these zeolites. However, they have channels with large-size aperture and also have very large spaces formed by intersecting channels. Therefore, their drawback is that some undesired molecules (high-boiling-point compounds) are formed in the intersections of the channels with large space, and the undesired molecules thus formed therein clogs the pores to cause activity depression.

Paragraph bridging pages 2 and 3:

Asl
Being different from these catalysts, mordenite-type zeolite has no intersection of large-pore channels, and few undesired large-size molecules can be formed therein. But the aperture size of the pores therein is not so large and it is not so effective for converting large-size molecules. Accordingly, the object of the present invention is to solve the prior art problems, precisely to provide a highly-active, selective and long-life method for converting aromatic compounds having a large molecular size, concretely, for converting at least one aromatic compound selected from (a) aromatic compounds having at least three substituents, (b) aromatic compounds having two substituents of which at least one is a halogen or has at least 2 carbon atoms, and (c) naphthalene or anthracene derivatives having substituent(s).

Page 3, first full paragraph:

To attain the subject matter as above, the method of the invention principally comprises contacting at least one aromatic compound with a zeolite-containing catalyst, wherein the zeolite is characterized in that;

As
(1) the minimum value of the pore aperture diameter of the major channels therein is larger than 0.65 nanometers (hereinafter referred to as nm), or the maximum value thereof is larger than 0.70 nm, and

(2) the major channels do not intersect any others with larger apertures than an oxygen 10-membered ring;

and the aromatic compounds are at least one compound selected from the group consisting of;

(a) aromatic compounds having at least three substituents,

(b) aromatic compounds having two substituents of which at least one is a halogen or has at least 2 carbon atoms, and

(c) naphthalene or anthracene derivatives having substituent(s).

Page 4, second full paragraph:

At The invention is described in detail hereinunder. Zeolite referred to herein is a crystalline microporous material which has the channels with uniform and molecular-level size pores, including crystalline aluminosilicate, crystalline metasilicate, crystalline metalloaluminosilicate, crystalline aluminophosphate, crystalline metalloaluminophosphate, and crystalline silicoaluminophosphate. The metasilicate and metalloaluminosilicate referred to herein are aluminosilicate derivatives in which aluminium is partly or entirely substituted with any other metals except aluminium, such as gallium, iron, titanium, boron, cobalt, chromium, etc. Similarly, metalloaluminophosphate indicates aluminophosphate derivatives in which aluminum or phosphorous is partly substituted with any other metals except itself.

Paragraph bridging pages 4 and 5:

Zeolite for use in the invention is not specifically defined for its composition, for which various compositions such as those mentioned above are adopted. However, its structure is indispensably defined as follows:

- An
- (1) the minimum value of the pore aperture diameter of the major channels therein is larger than 0.65 nm, or the maximum value thereof is larger than 0.70 nm, and
 - (2) the major channels do not intersect any others with larger apertures than an oxygen 10-membered ring.

Page 5, first full paragraph:

As In other words, the zeolite for use in the invention must satisfy the two requirements (1) and (2).

Paragraph bridging pages 5 and 6:

Ag The major channels of the zeolite referred to herein are meant to indicate the channels having the largest pore apertures in one zeolite structure. The pore size in zeolite, in general, is represented by reference to an oxygen n-membered ring in which n indicates the number

AG of oxygen atoms that constitute the pore aperture. For satisfying the requirement (1), the aperture size of the pores or channels must be at least equal to or larger than that of an oxygen 12-membered ring. Zeolite having channels of which the pore aperture size is larger than that of an oxygen 12-membered ring satisfies the requirement (1), but some others having the same do not. For example, mordenite-type zeolite, though having oxygen 12-membered ring pores(channels), does not satisfy the requirement (1), since the minimum value of the pore opening diameter of the pores therein is 0.65 nm and the maximum value thereof is 0.70 nm. On the other hand, SSZ-31 of which the structure has been recently clarified (R.F. Lobo et al.; J. Am. Chem. Soc., Vol. 119, pp. 3732-3744, 1997) satisfies the requirement (1), since the maximum value of the pore opening diameter of the pores therein is 0.86 nm and the minimum value thereof is 0.57 nm. Zeolite having oxygen 12-membered ring pores and satisfying the requirement (1) in regard to the pore aperture diameter, and zeolite having pores larger than 12-membered ring pores in regard to the pore aperture diameter, for example, zeolite having 14-membered ring pores are within the scope of zeolites for use in the invention.

Paragraph bridging pages 6 and 7:

ABZ
A10
Any one can know the sizes of pore apertures in zeolite of which the structure is known. Various types of zeolites of which the structure has been clarified, and the atomic configuration in such known different types of zeolites are described in Atlas of Zeolite Structure Types (W.M. Meier, D.H. Olson, Ch. Baerlocher, Zeolites, 17(1/2), 1996; Reference 1). In the section of Channels in Reference 1, they show the crystallographic free diameter. The free diameter values are based on any oxygen radius of 0.135 nm. In this shape, both the maximum value and the minimum value are shown for noncircular aperture. In this reference, the pore aperture is stereoscopically drawn as Fig. 1, and both its maximum value and the minimum value are given therein. The maximum value and the minimum value of the pore aperture referred to in the invention is just the values shown in the

A10
reference. For all types of zeolites clarified in Reference 1, their data of aperture size given in Reference 1 are referred to herein, irrespective of their composition, for judging their applicability to the invention (in fact, however, the pore size will vary, depending on the composition and the ambient temperature). For the others not shown in Reference 1 but disclosed in any other references such as journals and the like, their applicability to the invention will be judged in point of the pore aperture diameter from their structure disclosed in such other references.

Page 7, first full paragraph:

A11
For the types of zeolite not clarified in Reference 1 in reference to their pore aperture diameter, the maximum value and the minimum value of the pore aperture therein shall be determined in a simplified method such as that mentioned below. Based on the atomic configuration and the space group of zeolites shown in references, a crystal model is constructed. For this, the atomic configuration and the space group are preferably subject to a Rietveld refinement. The oxygen atoms constituting the pore aperture are combined with each other via a diagonal line drawn therebetween, and the maximum interatomic distance and the minimum interatomic distance are obtained (for this, the size of the oxygen atom itself is neglected, and the interatomic distance is obtained from the positional difference between the connected two oxygen atoms). From the maximum or minimum interatomic distance thus obtained, 0.27 nm (the diameter of one oxygen atom) is subtracted to give the maximum or minimum value of the pore opening size.

Paragraph bridging pages 7 and 8:

A12
Zeolite having a larger pore aperture is more preferred for use in the invention, as having a higher conversion activity and ensuring a higher reaction yield. Therefore, it is desirable that both the minimum value and the maximum value of the pore opening diameter of the pores in zeolite for use herein are larger than 0.70 nm. For the pore entrance of the major pores in zeolite for use herein, larger pores than oxygen 12-membered ring pores are

preferred to oxygen 12-membered ring pores. Though it is not clear, the reason may be because molecules could diffuse more rapidly in larger pores in the zeolite. If molecules diffuse slowly in pores in zeolite, their reaction will be retarded, or that is, the reaction activity in the pore of such zeolite is poor. In addition, if molecules diffuse slowly in pores in zeolite, the time for which they are contacted with zeolite shall be prolonged, thereby often inducing side reactions such as decomposition, and the reaction yield is lowered. Accordingly, enlarging the pore diameter in zeolite to promote the diffusion of molecules through the pores therein increases the conversion activity and the reaction yield. The uppermost limit of the pore diameter is not specifically defined, for which, however, it is desirable that the maximum value of the pore entrance diameter is at most 1.1 nm, more preferably at most 0.9 nm. Though it is not clear, the reason may be because too large pores, if any in the zeolite, will provide a site for large molecules to be formed therein, and high-boiling-point substances thus formed therein will clog the pores to shorten the catalyst life of the zeolite.

Page 9, first paragraph:

Zeolite for use in the invention must satisfy not only the requirement (1) indicating that the minimum value of the pore aperture diameter of the major channels therein is larger than 0.65 nm, or the maximum value thereof is larger than 0.70 nm, but also the requirement (2) indicating that the major channels do not intersect with any others that have larger apertures than oxygen 10-membered ring pores.

Paragraph bridging pages 9 and 10:

For its pore structure, zeolites have one-dimensional pore systems or poly-dimensional pore systems. Preferably, zeolite for use in the invention has essentially one-dimensional pores. The pore structure of the one-dimensional pore system is generally such that the major channels do not intersect with any other channels having pore sizes not smaller than oxygen 7-membered pores. Being different from it, the pore structure of poly-

dimensional pores is such that some major channels intersect some other channels with apertures that are not smaller than oxygen 7-membered ring pores. The invention is directed to conversion of aromatic compounds having a large molecular size, as will be described hereinunder, in which, therefore, zeolites having small pores that are not larger than oxygen 10-membered ring pores are substantially useless, since the pores therein are too small to be effective to perform the intended conversion. Accordingly, in the invention, it is recognized that the pore structures of zeolites in which the major channels intersect the other pores not larger than oxygen 10-membered ring pores are substantially one-dimensional, and zeolites of the type having such a one-dimensional pore structure are within the scope of zeolites for use in the invention. On the other hand, zeolite in which large channels intersect with each other, for example, β -type zeolite in which oxygen 12-membered ring pores intersect with each other shall have an extremely large space around the intersecting points, and undesirable reactions yielding high-molecular-weight substances will be inevitable in such a large space. The high-molecular-weight substances thus formed in the large space will clog the pores, whereby the activity of the zeolite will be lowered. This is the reason why one-dimensional pore-structured zeolites are preferred for use in the invention. Zeolite in which some major channels intersect some other channels having not larger apertures than oxygen 10-membered ring will have substantially no other open pores except the major channels, and this type of zeolite is adopted in accordance with the invention without problem. Preferably, however, in zeolite for use in the invention, the channels that may intersect the major channels do not have larger pores than an oxygen 10-membered ring.

Paragraph bridging pages 10 and 11:

The structure of zeolite for use in the invention is not specifically defined provided that it satisfies the requirements defined herein. With reference to the structural code of three letters indicated in Atlas of Zeolite Structure Types (W.M. Meier, D.H. Olson, Ch. Baerlocher, Zeolites, 17(1/2), 1996), concretely, the structure of zeolite for use in the

invention includes VFI, AET, AFI, AFR, AFS, ATS, BOG, BPH, DFO, GME, LTL, MAZ, MEI, OFF. In addition, it further includes CFI having large pores of which the pore entrance diameter is larger than that of oxygen 12-membered ring pores (M. Yoshikawa et al., Journal of Physical Chemistry, B, Vol. 102, pp. 7139-7147), and UTD-1 (R.F. Lobo et al., Journal of American Chemical Society, Vol. 119, pp. 8474-8484, 1997). Further, SSZ-31 mentioned above is also within the scope of zeolite for use in the invention. Of those, preferred for use herein are AFI, GME, LTL, MAZ, MEI, OFF, CFI, UTD-1, and SSZ-31; more preferred are CFI and UTD-1; even more preferred is CIT-5 zeolite. The structure type code of this zeolite is CFI.

Page 11, last paragraph:

Those types of zeolites may be natural ones or synthetic ones. Preferred is synthetic zeolite, as its composition can be controlled in any desired manner. Any known method can be adopted for producing it. For zeolite of which the structure is not clarified as yet, it will be impossible to identify the pore aperture diameter. If such large crystals of zeolite are prepared, the structure of the zeolite may be determined according to an M. Yoshikawa et al.'s method (Journal of Physical Chemistry, B, Vol. 102, pp. 7139-7147, 1998) or to a C.C. Freyhardt et al.'s method (Journal of American Chemical Society, Vol. 118, pp. 7299-7310, 1996).

Paragraph on page 12:

Specific examples of zeolite structures for use in the invention are mentioned hereinabove, and the composition of zeolite can be selected with reference to the intended reaction. For acid catalyst reaction, preferably used is any of crystalline aluminosilicate, trivalent metal-containing crystalline metallosilicate, and crystalline silicoaluminophosphate; more preferred are crystalline aluminosilicate and crystalline gallosilicate; and even more preferred is crystalline aluminosilicate. In crystalline aluminosilicate and metallosilicate for use in the invention, the ratio (by mol) of Si/(trivalent metal) is not specifically defined, but

A17 preferably falls between 5 and 500, more preferably between 5 and 200, even more preferably between 7 and 100. Zeolite having a smaller ratio of Si/(trivalent metal) will have a larger number of active sites, and is therefore preferred for use in the invention. On the other hand, zeolite having a larger ratio of Si/(trivalent metal) will be more hydrophobic, and is therefore preferred for conversion of organic compounds, since it can more easily absorb the reaction substrate. Zeolite having a well-balanced ratio of Si/(trivalent metal) in consideration of these facts may be suitably selected for use in the invention. For oxidation with hydrogen peroxide, preferred is crystalline titanosilicate. For hydroxylation with nitrogen suboxide, preferred is crystalline ferrosilicate.

Page 13, first paragraph:

A18 In case the zeolite for use in the invention has ion-exchanging sites, the sites may be ion-exchanged with any other different ions. In case the zeolite is used herein as an acid catalyst, in general, it is previously subjected to a few times of ion exchange treatment with an aqueous solution of an ammonium salt and then calcined to be an acid-type zeolite catalyst.

Page 13, last paragraph:

A19 For preferable use in the invention, the zeolite is formed. Zeolite alone may be formed, or may be granulated along with a binder such as alumina, clay, etc. For granulating it, for example, the zeolite is kneaded with a binder such as alumina or the like, then extruded out through an extruder, and rounded into granules by the use of an equipment like Marumerizer (a type of granulator, pelletizer or the like).

Page 14, first paragraph:

From the zeolite-containing catalyst, in general, crystal water existing therein and organic substances used in producing it and still remaining therein are removed before use.

A20 In general, it may be heated at 200 to 600°C, whereby crystal water and the organic substances can be almost completely removed from zeolite.

Page 14, second paragraph:

A21 The catalyst may contain metal. For example, it is desirable to prolong the catalyst life that the acid catalyst contains a noble metal, and the reaction is carried out in the presence of hydrogen. Though not clear, the reason may be because the catalyst could more readily receive protons and it could be prevented from coking. The noble metal that may be in the catalyst is not specifically defined, but rhenium is the best. The reason is because the catalyst containing rehenium will hardly undergo hydrogenolysis.

Page 15, first paragraph:

A22 The aromatic compounds in the invention are limited to those having a relatively large molecular size. For converting compounds having a small molecular size, such as xylene, toluene and others, conventional pentacyl-type zeolite (MFI) and mordenite-type zeolite (MOR) will give enough reaction property but they do not give enough reaction property for converting aromatic compounds having a large molecular size. The method of the invention is directed to the efficient conversion of such aromatic compounds having a relatively large molecular size. Aromatic compounds having a relatively large molecular size in the invention are at least one selected from:

- (a) aromatic compounds having at least three substituents,
- (b) aromatic compounds having two substituents of which at least one is a halogen or has at least 2 carbon atoms, and
- (c) naphthalene or anthracene derivatives having substituent(s).

Page 20, first full paragraph:

A23 Ludox HS-30 (from DuPont) was used as a silica source; aluminium nitrate 9-hydrate (from Nakarai Tesq) was used as an aluminium source; and anhydrous lithium hydroxide (from Kishida Chemical) was used as a lithium source. From these, prepared was a mixture having a compositional formula, $\text{SiO}_2:0.1\text{LiOH}:0.2\text{MeSPA}\text{OH}:0.01\text{Al}(\text{NO}_3)_3:40\text{H}_2\text{O}$ (by mol).

Page 20, second full paragraph:

A24 The mixture was stirred for 2 hours, and then heated in a Teflon lined autoclave at 175°C for 9 days. The resulting zeolite was taken out through filtration, washed with water, dried at 100°C and analyzed through X-ray diffractionmetry, by which it was identified as CIT-5. According to the reference (M. Yoshikawa et al's Journal of Physical Chemicatry, B, Vol. 102, pp. 7139-7147, 1998), the zeolite CIT-5 contains oxygen 14-membered ring pores, and the pore aperture is nearly circular and has a diameter of about 0.73 nm. In this, the major channels do not intersect any other pores.

Page 22, first full paragraph:

A25 The relative ratio of 2,4-DCT conversion per one active site of the catalyst used was as follows: The number of the active sites in each catalyst was determined from the amount of aluminium in zeolite. The degree of conversion was determined from the weight loss of 2,4-DCT before and after the reaction.